

## Temporal-spatial pattern of organic carbon sequestration by Chinese lakes since 1850

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### Abstract

In the last century, lakes in China have been subject to forcing by climate change, intensification of agriculture, and urban expansion, though their effects on lake OC sequestration are poorly understood. We compiled dry mass and OC burial rates from 82 <sup>210</sup>Pb-dated lake sediment records in China. The average post-1950 focusing-corrected lake mass accumulation rate (MAR<sub>FC</sub>) of  $256 \pm 56 \text{ g m}^{-2} \text{ yr}^{-1}$  (median  $\pm$  SE) and focusing-corrected OC accumulation rate (CAR<sub>FC</sub>) of  $8 \pm 3 \text{ g C m}^{-2} \text{ yr}^{-1}$  were significantly higher than the 1850–1900 rates ( $p < 0.05$ ). However, the magnitude of increase in CAR<sub>FC</sub> was most marked in the subtropical lakes of the East Plain (EP) and on the Yunnan-Guizhou Plateau (YG), where the post-1950 CAR<sub>FC</sub> was about three times that of the 1850–1900 ( $p < 0.05$ ), due to the agricultural intensification and urban expansion in recent decades. Moreover, MAR<sub>FC</sub> was significantly higher in the EP than that on the Mongolia-Xinjiang Plateau (MX) for all time periods ( $p < 0.05$ ). Lake CAR<sub>FC</sub> in YG was significantly higher than rates in the Qinghai-Tibetan Plateau (QTP) for the post-1950 and MX for the 1850–1900 ( $p < 0.05$ ). Regression analyses showed that the controls on lake CAR<sub>FC</sub> varied among regions, with catchment climate variables the most important regulators in MX, Northeast China, and QTP, but the in-lake nutrient concentrations were more important in YG and EP ( $p < 0.05$ ). The results from this study show how modern limnic OC sequestration has changed with human disturbance and climate change in China.

### Highlights

- First estimation of focusing-corrected contemporary lake organic carbon accumulation rate (CAR<sub>FC</sub>) at a national scale
- Lake CAR<sub>FC</sub> for post-1950 was significantly higher than that of 1850–1900

- Lake CAR<sub>FC</sub> and its increase rate varied significantly among different regions
- The controls on lake CAR<sub>FC</sub> varied among different regions
- Evidence of agricultural intensification, urban expansion, and climate-regulated lake organic carbon accumulation variability

The importance of lakes to the global carbon (C) cycle is related to their capacity to process organic matter (OM) derived from surrounding catchments as well as that produced by aquatic primary production (Tranvik et al. 2009). Lakes emit large amounts of C to the atmosphere, with an estimated release of  $320 \text{ Tg C yr}^{-1}$  ( $1 \text{ Tg} = 10^{12} \text{ g}$ ) as CO<sub>2</sub>

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Additional Supporting Information may be found in the online version of this article.

(Raymond et al. 2013) and  $48 \text{ Tg C yr}^{-1}$  as  $\text{CH}_4$  (Bastviken et al. 2004). For most lakes, especially for those with dissolved organic carbon (OC) concentrations greater than  $10 \text{ mg C L}^{-1}$ , ecosystem respiration is higher than production (Algesten et al. 2004). However, lakes are also long-term C sinks, with an estimated total C burial of  $\sim 42 \text{ Tg C yr}^{-1}$  (Dean and Gorham 1998). Approximately 25% (ranging from 11% to 46%) of the combined OC inputs from terrestrial sources and in situ production is estimated to be permanently buried by lakes (Sobek et al. 2009), emphasizing the relatively high OC burial rate of lakes, comparable to (or even higher than) that deposited in marine sediments (Sobek et al. 2009) and terrestrial soils (Gudasz et al. 2010).

In the past century, climate change and human activities have exerted significant influence on the C dynamics of lakes. On the one hand, climate warming and anthropogenic disturbance can promote lake OC burial by increasing the amount of OC produced within lakes (Tranvik et al. 2009) and/or from terrestrial ecosystems (Canham et al. 2004; Sobek et al. 2007; Tranvik et al. 2009). Moreover, warming and eutrophication associated with human activities can enhance OC preservation since they lead to an increased hypolimnetic oxygen depletion rate (Radbourne et al. 2017). The resultant anoxia reduces mineralization rates at depth in the hypolimnion and at the sediment surface in the profundal zone (Radbourne et al. 2017). On the other hand, these processes (temperature, eutrophication) may enhance OC mineralization (Bastviken et al. 2004; Song et al. 2012), because they stimulate microbial activity.

The response of the lake OC burial rate to natural environmental forcing and anthropogenic disturbance is a function of the imbalance between the increase of OC input from in situ production and/or from terrestrial ecosystems and that of OC losses via mineralization and outwash. The long-term carbon accumulation rate (CAR) was estimated at  $6 \text{ g C m}^{-2} \text{ yr}^{-1}$  in southwest Greenland (Anderson et al. 2009),  $5.6 \text{ g C m}^{-2} \text{ yr}^{-1}$  in Europe (Kastowski et al. 2011),  $3.8 \text{ g C m}^{-2} \text{ yr}^{-1}$  in northern Quebec (Ferland et al. 2012), and  $\sim 7.7 \text{ g C m}^{-2} \text{ yr}^{-1}$  in China (Wang et al. 2015), although most of these rates are not focusing corrected. However, human activities (notably land-cover change and increased nutrient loads) have increased lake OC burial rate significantly, with a mean modern CAR of  $60 \text{ g C m}^{-2} \text{ yr}^{-1}$  for 90 agriculturally impacted European lakes (Anderson et al. 2014) and  $\sim 70 \text{ g C m}^{-2} \text{ yr}^{-1}$  for lakes in Minnesota, U.S.A. (Anderson et al. 2013). Atmospheric deposition of reactive nitrogen was inferred to have increased the contemporary OC burial rates by fivefold (to a mean of  $\sim 15 \text{ g C m}^{-2} \text{ yr}^{-1}$ ) in northern lakes unaffected by direct human activity during the last century (Heathcote et al. 2015). Therefore, it is important to have better estimates of recent C burial by lakes that are subject to the pressure of anthropogenic impact and climate change (Battin et al. 2009).

Over the last century, besides climate change, anthropogenic disturbance has also affected lakes across the different regions of China, including rapid urbanization, extensive agriculture, and intensive aquaculture, causing eutrophication (Qin and Zhu 2006) and increasing the terrestrially-derived OM input (Xue and Yao 2011). Although the C burial by China's lakes has been established as an important aspect of the regional terrestrial C cycle (Dong et al. 2012; Wang et al. 2015), it is still not fully understood how OC burial rate of lakes in China changed over time, what the contemporary burial rates are, nor their response to climate change and widespread regional human activity, limiting our understanding of the role of lakes in climate regulation. Moreover, lakes in China vary regionally reflecting not only the natural environment conditions (climate, geology), including their origin and long-term development, but also the intensity and type of anthropogenic disturbance (Wang and Dou 1998). This regional variability can influence the modern OC accumulation rate and the magnitude of its increase across the country.

In this study, data from 82 lakes in China were compiled, (1) to quantify the OC burial rate since 1850 at regional scale, (2) to determine the extent to which the lake OC burial rates have increased over the past 150 yr among the different regions in China, and (3) to identify the mechanisms underlying the temporal and spatial variations in lake OC burial rate. It was hypothesized that first, the modern OC accumulation rate should be enhanced by either climate warming and/or agricultural disturbance for lakes in China as a whole; and second, regional differences exist in both OC accumulation rate and the degree of increase during the 20<sup>th</sup> century, with higher OC accumulation rate in regions where the climate is warmer and wetter, agricultural land-use more intensive and urbanization more extensive, when compared with other regions.

## Data sources and analysis methods

### Lake sites

Lakes cover an area of  $\sim 91,019 \text{ km}^2$  in China and are located in five main regions: East Plain (EP) of subtropical China, Yunnan-Guizhou Plateau in southwest China (YG), Mongolia-Xinjiang Plateau (MX), the Qinghai-Tibetan Plateau (QTP), and Northeast China (NEC) (Fig. 1) (Wang and Dou 1998). Among the five major lake regions in China, the QTP and MX are located in the mainland hinterland with an arid and semi-arid climate, and most of the lakes are saline (Wang and Dou 1998). Lakes in YG, NEC, and EP are located in the Asian monsoon region with a humid climate and most of the lakes have outflows and are fresh (Wang and Dou 1998). In recent decades, with the rapid development of agriculture and industry as well as massive urbanization, lake catchments have been greatly altered, causing significant environmental problems for Chinese lakes including

ecosystem degradation, eutrophication, and reduction in flood storage capacity (Yang et al. 2010). As with the lakes themselves, these problems also show regional differences, with the most serious impacts found in EP, followed by YG and NEC, then in MX and QTP (Wang and Dou 1998).

The data from the 82 lakes used in this study were mainly derived from published sources (*see* Supporting Information). Due to the vast size of the country, lakes in China differ greatly in terms of local climate, catchment topography, area, water depth, and nutrient concentration (Supporting Information Table S1). The lakes are distributed across diverse climate zones, from humid and warm regions to cold and dry zones as well as from low altitude coastal plains to alpine plateau basins. They span a latitudinal range from 24.3°N to 48.7°N, a longitudinal range from 80°E to 133°E and an altitudinal gradient from 3 m to 5030 m above sea level (Supporting Information Table S1). The mean annual air temperature ranges from around  $-8^{\circ}\text{C}$  to  $+24^{\circ}\text{C}$  and the annual precipitation from  $\sim 24$  mm to 2000 mm (Supporting Information Table S1). Lake size ranges from  $\sim 1$  km<sup>2</sup> to 4340 km<sup>2</sup>, with only six lakes larger than 1000 km<sup>2</sup> in EP, NEC, MX, and QTP. The lakes also cover a wide range of water depths (mean depths are  $\sim 1$ –87 m; maximum depths ranging from  $\sim 2$  m to 157 m) and nutrient levels. Most lakes in EP are shallow (water depth  $< 5$  m) and nutrient-rich, but the lakes in QTP are deep and oligotrophic (Supporting Information Table S1).

### Data collection and analysis

The details of the coring and processing methods can be found in the original publications (*see* Supporting Information), and are only described briefly here. A piston corer was mainly used to collect the sediment cores and in general the upper 30 cm was sampled at 0.5–2 cm intervals for the analysis of <sup>137</sup>Cs and <sup>210</sup>Pb (Appleby 2001) and radiometric chronologies were calculated from the <sup>210</sup>Pb records using the constant rate of supply model (Appleby et al. 1986). The sediment accumulation rate was multiplied by dry bulk density (DBD) to generate the mass accumulation rate (MAR); MAR was then multiplied by the sediment OC content to obtain C accumulation rate (CAR). The sediment inorganic C fraction was not counted in this study due to the limited data available although lakes in the dry regions may have high inorganic C content (Li et al. 2017). For most sites, the OC content was determined by combustion using an Elemental Analyzer with reference to standard samples and for those lacking OC values (Chaiwopu, Sugan Lake, Gahai Lake, and Wudalianchi), it was estimated by converting loss-on-ignition, which was determined using standard methods and converted by a correction factor (0.469) (Dean 1974). DBD was measured on 1 cm<sup>3</sup> subsamples after drying in an oven for 16 h at 105°C, following the standard method (Rausch and Heinemann 1984). Direct measurements of DBD were not available for nineteen lakes (Supporting Information Table S1). For these lakes, it was

generated based on the following empirical relationship with OC content. In case of high OC content ( $> 6\%$ ), the relationship:  $\text{DBD} = 1.665 * (\text{OC})^{-0.887}$  given by (Dean and Gorham 1998) was used. In case of low OC content ( $\leq 6\%$ ), the DBD for sediments was calculated using the formula:  $\text{DBD} = 1.776 - 0.363 * \ln(10 * \text{OC})$  (Avnimelech et al. 2001). For both methods, the OC content is reported as percent C and DBD as g cm<sup>-3</sup>.

It is well known that sediment accumulates nonuniformly in lakes (Brezonik and Engstrom 1998; Engstrom et al. 2007). Therefore, a single sediment core from a lake is unlikely to provide an unbiased estimate of the average OC burial rate (Engstrom and Rose 2013). Though multiple cores from a single lake can be used to improve the estimation of the mean lake OC burial rate, such an extremely labor intensive approach cannot be realistically expected for regional studies where multiple sites are sampled. An alternative approach is to estimate the sediment focusing factor using the ratio of the mean sediment <sup>210</sup>Pb flux to the regional atmospheric <sup>210</sup>Pb flux (Engstrom and Rose 2013). In this study, multiple-core data were available for 17 lakes (two cores for seven sites, three cores for four sites, four cores for two sites, five and six cores for one site, seven cores for two sites) (Supporting Information Fig. S2), allowing the basin average CAR to be estimate directly; for those lakes with less than three cores (Supporting Information Figs. S1, S2), we used the focusing factor to adjust the CAR estimate. <sup>210</sup>Pb flux data were not available for 19 lakes with only single core, and average focusing factor of studied lakes in each region was used to estimate their focusing corrected MAR (MAR<sub>FC</sub>) and focusing corrected CAR (CAR<sub>FC</sub>). Those lakes without <sup>210</sup>Pb flux data were excluded from the regional comparisons except for mass and OC burial increase magnitude calculations. Although the atmospheric <sup>210</sup>Pb flux is not well prescribed for each site, it was assumed to be proportional to rainfall and calculated according to the relationship of 100 Bq kg<sup>-1</sup> per year per 1000 mm of precipitation (Appleby 2001).

Post-deposition mineralization is another issue that needs to be addressed when estimating OC burial rate in lakes. Although sediment OM degradation can take place over centuries or thousands of years, the degradation rate decreases over time (Rothman and Forney 2007). A study that tracked OC over 27 yr using annual varves indicated that  $\sim 87\%$  of sedimentary-C loss occurred within the first 5 yr and that the older sediments lost less than 1% of sediment C per year (Gälman et al. 2008). In order to eliminate the effect of post-depositional mineralization on the temporal variability of CAR, sediments younger than 10 yr were excluded from calculations, as in earlier studies (Anderson et al. 2014; Heathcote et al. 2015). The MAR<sub>FC</sub> and CAR<sub>FC</sub> of each lake was calculated for three periods (1850–1900, 1900–1950, and post-1950 [referred to 1950–2000]) by averaging the rates within each period. We used the median of MAR<sub>FC</sub> and

CAR<sub>FC</sub> values from the study lakes within a given region to generate the regional mean, and the error bar (standard error) represents the differences in the rates among different lakes within a region. One-sided paired *t*-tests were used to make the statistical comparisons of MAR<sub>FC</sub> and CAR<sub>FC</sub> between periods (SAS 9.1 Institute, Cary, North Carolina, U.S.A.). One-way analysis of variance was used to determine whether the mean MAR<sub>FC</sub> and CAR<sub>FC</sub> differed significantly among regions for different periods. Multiple comparisons among means were obtained by Duncan's multiple comparison procedure at the significance level of 0.05 (SAS 9.1 Institute, Cary, North Carolina, U.S.A.).

Linear regression analyses between the post-1950 CAR<sub>FC</sub> and epilimnetic nutrient concentrations (total nitrogen [TN] and total phosphorus [TP]), and lake physical catchment characteristics (i.e., lake surface area, lake water depth, mean annual precipitation, and temperature), as well as the sediment C : N ratio were undertaken. All the data were log-transformed before analysis to confirm their normality. Moreover, stepwise multivariable regression analyses were conducted to determine the relative importance of these variables in controlling the spatial variation in lake CAR<sub>FC</sub> for post-1950 in the different regions. These analyses, undertaken with the statistical program SAS v9.1, were applied to every lake that had data for both CAR<sub>FC</sub> and the relevant lake and catchment characteristics. Lake area, maximum water depth, annual temperature, and precipitation data were available for most lakes. In contrast, TN and TP concentration data were available for only 33 lakes and were not available for most lakes in QTP and NEC. The C : N ratio was available for 26 lakes. The correlation coefficient between CAR<sub>FC</sub> and MAR<sub>FC</sub> was also determined. The study period was divided into three representative periods: 1850–1900 was considered the period with low disturbance; 1900–1950 a transitory period with increasing human population and industrialization, and post-1950 was considered as the modern period undergoing intensive pressure from human population, agriculture, and industrialization.

For QTP, MX, and NEC, the results of regression analysis between CAR<sub>FC</sub> and lake size (see "Results" section) were applied to each lake and the annual regional focusing-corrected OC sequestration capacity was calculated by summing the CAR<sub>FC</sub> for each lake within a given region. For EP and YG, the annual regional focusing-corrected OC sequestration capacity was calculated by simple extrapolation of median CAR<sub>FC</sub> of study lakes since no significant regression relationships between CAR<sub>FC</sub> and lake size were found (see "Results" section). Data on lake size and location were obtained from a former national survey of Chinese lakes (The Ministry of Water Resources of the People's Republic of China 1998; Wang and Dou 1998) and the data available in the public domain are given in Supporting Information Table S2.

## Results

The MAR<sub>FC</sub> for all study individual lakes (all time periods considered) ranged from  $< 50 \text{ g m}^{-2} \text{ yr}^{-1}$  to  $> 1500 \text{ g m}^{-2} \text{ yr}^{-1}$  (Table 1). CAR<sub>FC</sub> ranged from  $< 1 \text{ g C m}^{-2} \text{ yr}^{-1}$  to  $> 50 \text{ g C m}^{-2} \text{ yr}^{-1}$  for all study lakes (Table 1). The post-1950 MAR<sub>FC</sub> of all study lakes in China averaged  $256 \pm 56 \text{ g m}^{-2} \text{ yr}^{-1}$  (median  $\pm$  SE) and  $165 \pm 55 \text{ g m}^{-2} \text{ yr}^{-1}$  for the period 1900–1950, significantly higher than that of  $136 \pm 26 \text{ g m}^{-2} \text{ yr}^{-1}$  for 1850–1900 ( $p < 0.05$ ) (Fig. 2a). The post-1950 lake CAR<sub>FC</sub> averaged  $8 \pm 3 \text{ g C m}^{-2} \text{ yr}^{-1}$ , which is significantly higher than that for 1850–1900 ( $4 \pm 1 \text{ g C m}^{-2} \text{ yr}^{-1}$ ) ( $p < 0.05$ ) (Fig. 2b).

The MAR<sub>FC</sub> and CAR<sub>FC</sub> of the study lakes for each period showed significant differences among regions, as did the magnitude of increase (Figs. 2, 3). The MAR<sub>FC</sub> of the study lakes in EP was significantly higher than that in MX, QTP, and NEC for all time periods ( $p < 0.05$ ) (Fig. 2a). The MAR<sub>FC</sub> of the study lakes in YG was significantly higher than that in MX for all time periods but that in QTP and NEC for only post-1950 ( $p < 0.05$ ) (Fig. 2a). The MAR<sub>FC</sub> of the study lakes in QTP and NEC was significantly higher than that in MX for the period 1900–1950 ( $p < 0.05$ ) (Fig. 2a). CAR<sub>FC</sub> of the study lakes in YG was higher than that in MX and QTP for the period post-1950 ( $p < 0.05$ ) but only higher than that in MX for 1850–1900 ( $p < 0.05$ ) (Fig. 2b). CAR<sub>FC</sub> of the study lakes in NEC was significantly higher than that in MX and QTP for the period 1900–1950 but only higher than that in MX for 1850–1900 (Fig. 2b).

MAR<sub>FC</sub> of the study lakes was not significantly different among periods across all regions except for YG and EP, where the post-1950 MAR<sub>FC</sub> was significantly higher than that for the period 1850–1900 ( $p < 0.05$ ) (Fig. 2a). The CAR<sub>FC</sub> of the study lakes for the post-1950 and 1950–1900 periods was significantly higher than that for 1850–1900 for lakes in MX, YG, and EP ( $p < 0.05$ ), but no significant variation was found in NEC and QTP (Fig. 2b). Also, the CAR<sub>FC</sub> of the study lakes for 1950–1900 was significantly higher than that for 1850–1900 in EP (Fig. 2b). Moreover, the CAR<sub>FC</sub> of the study lakes increased by  $\sim 11 \pm 4 \text{ g C m}^{-2} \text{ yr}^{-1}$  in EP,  $\sim 24 \pm 10 \text{ g C m}^{-2} \text{ yr}^{-1}$  in YG, and  $\sim 15 \pm 10 \text{ g C m}^{-2} \text{ yr}^{-1}$  in NEC from 1850–1900 to post-1950 (Fig. 3a). Although the differences among time periods varied by region (Fig. 3a), the temporal pattern of CAR<sub>FC</sub> indicated an increasing trend for lakes in China as a whole (Fig. 3b).

Regression analyses showed that lake CAR<sub>FC</sub> was correlated with different variables among the study regions (Fig. 4). The post-1950 CAR<sub>FC</sub> was positively correlated with the in-lake TP in YG, but with annual precipitation in the other regions (Fig. 4; Supporting Information Table S3). The post-1950 CAR<sub>FC</sub> in QTP was also positively related to the mean annual air temperature (Fig. 4c; Supporting Information Table S3). The post-1950 CAR<sub>FC</sub> for all lakes was positively correlated with the in-lake water TP concentration (Fig. 4a; Supporting Information

**Table 1.** MAR<sub>FC</sub> and CAR<sub>FC</sub> for different time periods of the study lakes.

Region	Site name	Focusing corrected mass and OC accumulation rate (g m <sup>-2</sup> yr <sup>-1</sup> )					
		Post-1950		1900–1950		1850–1900	
		MAR <sub>FC</sub>	CAR <sub>FC</sub>	MAR <sub>FC</sub>	CAR <sub>FC</sub>	MAR <sub>FC</sub>	CAR <sub>FC</sub>
EP	Chaohu	1047	10	605	8	83	6
EP	Dianshan	387	4	326	1	264	1
EP	Guchenghu	687	15	395	7	—	2
EP	Nanyi	1452	11	—	—	—	—
EP	Shijiu	640	27	1099	11	478	11
EP	Taibai	456	9	1119	20	561	5
EP	Taihu	—	56	—	7	—	3
EP	Wushan	291	10	397	10	140	4
EP	Yangcheng	1410	32	1139	14	625	21
EP	Zhangdu	—	9	—	16	—	8
EP	Poyang	6262	16	3239	6	1366	3
EP	Baiyangdian	354	6	409	5	391	5
EP	Chibaniuehu	892	9	—	—	—	—
EP	Donghu	817	14	714	14	—	—
EP	Dongping	354	8	352	13	203	8
EP	Dongtinghu	62	2	—	—	—	—
EP	Honghu	1599	60	1617	25	—	—
EP	Longgan	—	14	—	1	—	0.3
EP	Liangzihu	186	12	54	2	—	—
EP	Shitang	737	15	588	10	—	—
EP	Wanghu	251	20	147	13	—	—
EP	Yunzhonghu	556	19	—	—	—	—
MX	Aibi	12	1	17	1	11	1
MX	Bosten	46	0.4	21	0.2	12	0.1
MX	Jili	37	1	36	1	0.0	0.5
MX	Wuliangsu Hai	335	2	457	2	264	1
MX	Aerjilin	352	11	343	8	—	—
MX	Badan	160	1	109	1	106	0.5
MX	Buluntuohai	28	1	22	1	—	—
MX	Chaonaqiu	77	4	67	5	41	2
MX	Chenpu	54	1	22	0.2	21	0.1
MX	Chaiwopu	193	5	202	5	217	4
MX	Dundejilin	903	28	496	12	356	9
MX	Gonghai	187	13	66	4	—	—
MX	Hongjiannao	639	9	1378	7	—	—
MX	Shuanghaizi	204	6	142	3	—	—
MX	Sailimuhu	76	3	34	1	38	1
MX	Tiewake	15	1	13	0.5	—	—
MX	Yindeertu	58	2	135	4	168	4
MX	Zhunaogeqi	166	5	51	1	32	1
NEC	Lianhuan	262	7	193	5	136	4
NEC	Wudalianchi3	161	6	—	—	—	—
NEC	Wudalianchi5	132	5	—	—	—	—
NEC	Yueliang	778	119	910	139	180	28
NEC	Sihailongwan	193	13	163	11	147	10
NEC	Xiaolongwan	189	13	160	11	144	10

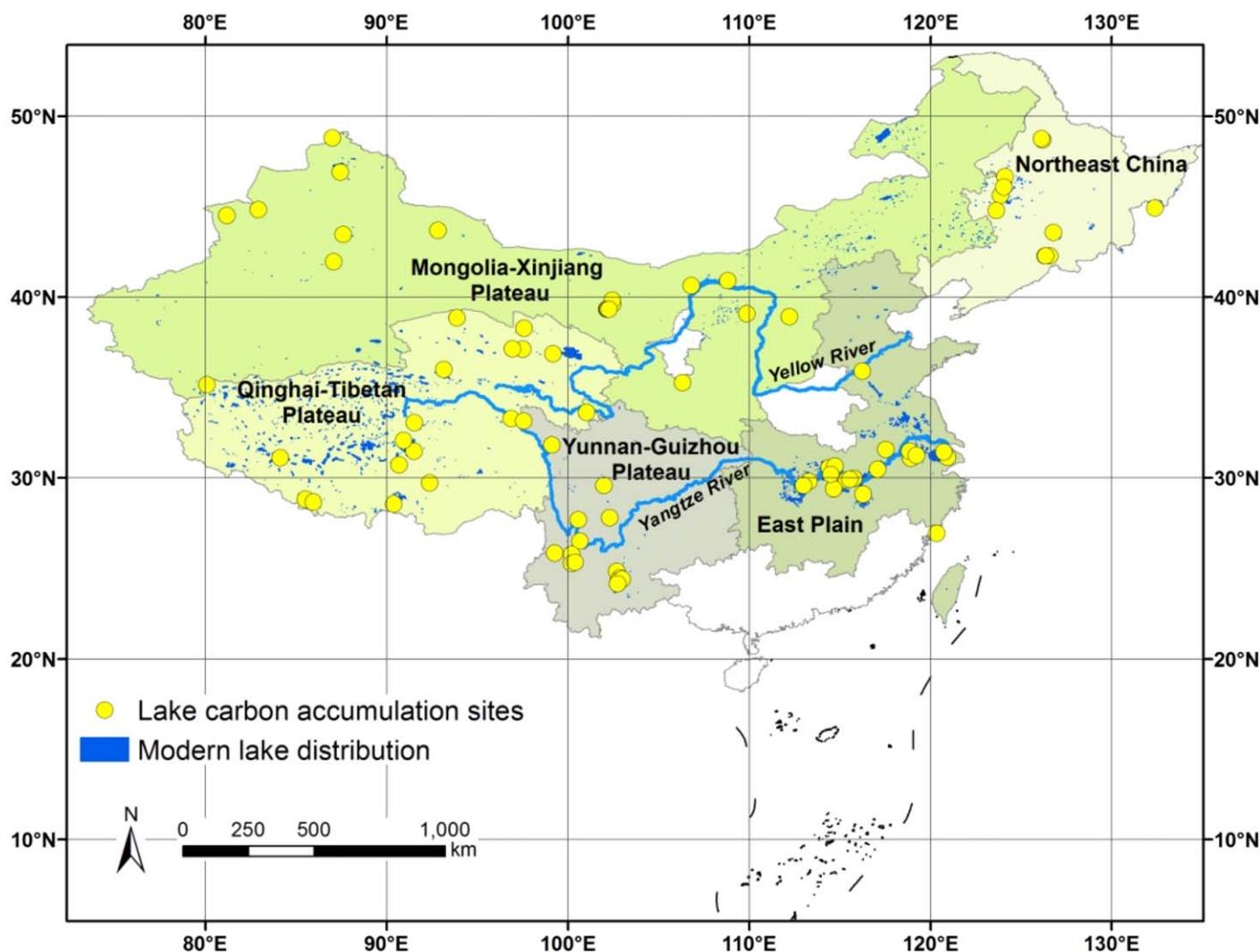
**TABLE 1.** Continued

Region	Site name	Focusing corrected mass and OC accumulation rate ( $\text{g m}^{-2} \text{yr}^{-1}$ )					
		Post-1950		1900–1950		1850–1900	
		MAR <sub>FC</sub>	CAR <sub>FC</sub>	MAR <sub>FC</sub>	CAR <sub>FC</sub>	MAR <sub>FC</sub>	CAR <sub>FC</sub>
NEC	Nashinapao	301	4	387	3	308	2
NEC	Erlongwan	—	24	—	9	—	5
NEC	Xidanghai	125	1	49	0.3	28	0.2
NEC	Xingkaihu	357	11	282	12	73	3
QTP	Cuo'e	1361	14	845	9	475	5
QTP	Nam Co	449	5	338	4	—	—
QTP	Qinghai	129	4	120	4	108	3
QTP	Sugan	195	12	152	10	115	7
QTP	Tarocuo	515	12	516	12	473	13
QTP	Xinluhai	619	15	724	17	—	—
QTP	Gahai	112	4	—	—	—	—
QTP	Pumyum Co	152	3	167	3	136	2
QTP	Hala	97	3	119	3	95	4
QTP	Zigetang	88	2	74	1	94	2
QTP	Hongshan	140	2	153	2	172	2
QTP	Caohaizi	59	1	18	0.4	19	0.4
QTP	Cuona	904	18	186	4	—	—
QTP	Dongerwuka	62	1	51	1	51	1
QTP	Kekuke	210	10	72	4	21	1
QTP	Kemen Co	195	21	143	11	—	—
QTP	Nir'pa Co	0.3	0.01	0.4	0.01	0.4	0.01
QTP	Peku Co	322	4	—	—	—	—
QTP	Tuosuhu	40	1	53	1	18	0.3
QTP	Yanghu	828	16	159	3	113	2
YG	Chenghai	1400	24	—	—	—	—
YG	Dianchi	973	61	863	27	379	7
YG	Fuxian	1001	81	907	44	321	16
YG	Qingshui	310	5	227	2	—	—
YG	Erhai	136	4	138	2	139	2
YG	Heihai	326	8	332	8	279	7
YG	Lugu	319	7	224	6	136	2
YG	Qionghai	523	7	153	0.4	—	—
YG	Qiluhu	416	64	—	—	—	—
YG	Xingyun	675	19	815	12	458	8
YG	Yunlongtianchi	55	2	22	0.5	—	—
YG	Yangzhonghu	1011	40	552	12	269	6

CAR, organic carbon accumulation rate; EP, East Plain of subtropical China; FC, focusing-corrected; MAR, mass accumulation rate; MX, Mongolia-Xinjiang Plateau; NEC, Northeast China; QTP, Qinghai-Tibetan Plateau; YG, Yunnan-Guizhou Plateau in southwest China.

Table S3) and the mean annual precipitation (Fig. 4b; Supporting Information Table S3), but not significantly related with the mean annual air temperature (Fig. 4c; Supporting Information Table S3). In addition, CAR<sub>FC</sub> was found to vary positively with the C : N ratio of lake sediments for all regions except for QTP and its corresponding MAR<sub>FC</sub> for all regions (Fig. 5; Supporting Information Table S3). Moreover, results of multivariable regression analysis suggested that lake CAR<sub>FC</sub>

for post-1950 was positively related to the mean annual precipitation and air temperature in MX, QTP, and NEC, but with TP in YG. For EP, the lake CAR<sub>FC</sub> for post-1950 was positively correlated with TN, pH, and annual precipitation, but negatively with air temperature (Table 2). Lake surface area was found to be significantly related to CAR<sub>FC</sub> for all periods in MX, QTP, and NEC, but not in EP and YG (Fig. 6; Supporting Information Table S4).



**Fig. 1.** Location of the lake sites in China used in this study. The lake regions are EP of subtropical China, YG, MX, the QTP, and NEC. [Color figure can be viewed at [wileyonlinelibrary.com](http://wileyonlinelibrary.com)]

The total OC burial rate by lakes in China as a whole was  $0.89 \text{ Tg yr}^{-1}$  for post-1950,  $0.59 \text{ Tg yr}^{-1}$  for 1900–1950, and  $0.34 \text{ Tg yr}^{-1}$  for 1850–1900 (Table 3). Overall, the estimated post-1950 total OC burial by lakes in China was  $44.38 \text{ Tg}$ ,  $29.67 \text{ Tg}$  for the period 1900–1950, and  $16.79 \text{ Tg}$  for the period 1850–1900, with more than 80% contributed by the lakes in EP and QTP (Table 3).

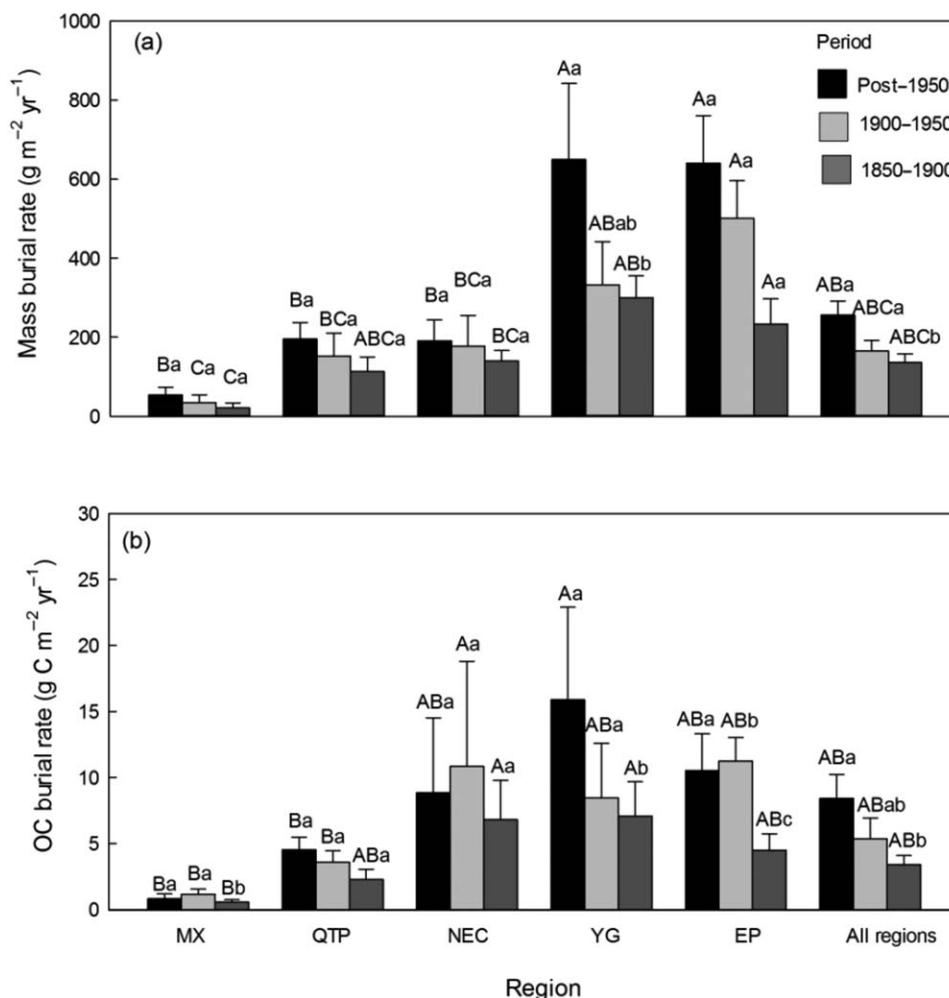
## Discussion

### Spatiotemporal patterns of OC accumulation of lakes in China

For study lakes in China as a whole, there were significant increases in mass and OC burial rate over the past 100 yr (Figs. 2, 3). This observation supports the hypothesis that human activities during the past century have increased the bulk sediment and OC burial of lakes in China. The post-

1950 OC burial rate ( $\sim 8.4 \pm 3.1 \text{ g C m}^{-2} \text{ yr}^{-1}$ ) was estimated to be about 2.5 times that for 1850–1900 ( $3.4 \pm 1.0 \text{ g C m}^{-2} \text{ yr}^{-1}$ ) (Fig. 2). Similarly, earlier studies found that the OC burial rate increased by a factor of  $\sim 3$  in European lakes (Anderson et al. 2014) and five times in lakes of North America (Heathcote et al. 2015), due to direct or indirect human influences on lakes and their catchments. Similarly, cultural disturbance also explains the high contemporary OC burial rate in Chinese lakes.

As the high recent OC burial rate of lakes may partly be due to incomplete mineralization of recently deposited sediments (Teranes and Bernasconi 2000; Gälman et al. 2008), the uppermost sediment (i.e., the most recent  $\sim 10 \text{ yr}$ ) were excluded when estimating the  $\text{CAR}_{\text{FC}}$  as was done in the other studies cited previously. In addition, the effect of long-term decomposition of lake sediments was considered by using a degradation model (Zimmerman and Canuel

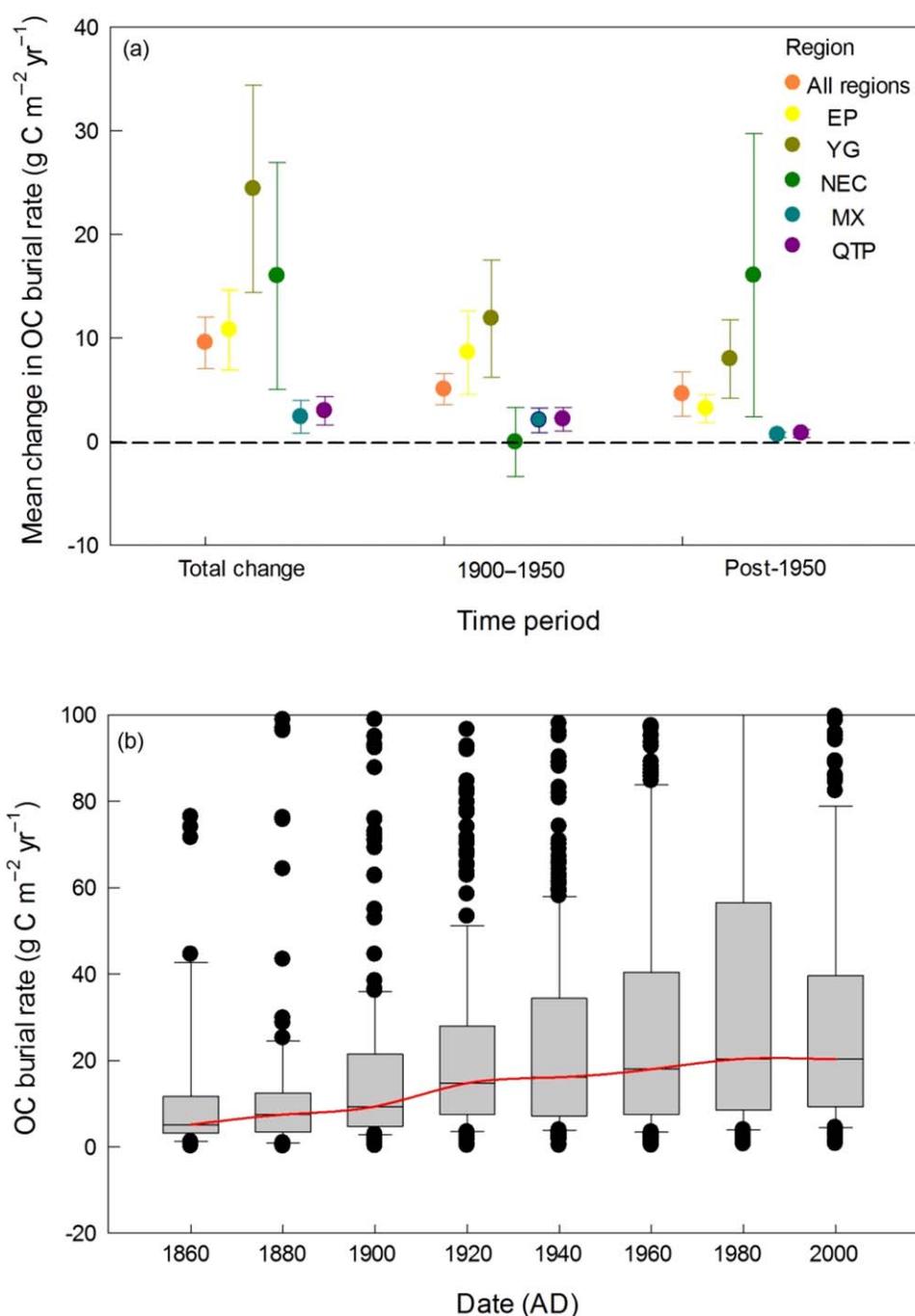


**Fig. 2.** Mass and OC burial rate (median  $\pm$  SE) of lakes in different regions of China during three time periods: (a,b) post-1950, 1900–1950, and 1850–1900. Different lowercase letters on the bars indicates a significant difference in mass and OC burial rate of each region for the different periods, and varying uppercase letters indicate significant differences in mass and OC accumulation rate for each time period among regions.

2002) and no change in the temporary pattern of OC burial rate was found. Therefore, post-depositional degradation of, or incomplete mineralization of lake sediments, are the primary factors explaining the observed changes or the high contemporary OC burial rates.

Contradicting the hypothesis that lakes in regions with a warm and wet climate, as well as pronounced anthropogenic activity, should have higher OC burial rates, the  $CAR_{FC}$  of lakes in EP was not significantly different with that of lakes in the other regions, though lake  $MAR_{FC}$  in EP was significantly higher than that in MX (Fig. 2). Although the post-1950  $CAR_{FC}$  of agriculturally-impacted lakes in EP was low ( $10.5 \pm 4.8 \text{ g C m}^{-2} \text{ yr}^{-1}$ ), the magnitude of increase relative to the 1850–1900 burial rate was large, suggesting that although the background OC burial rate was low, the accumulation rate was significantly increased by the recent intensification of agriculture, in line with our primary hypothesis. The low  $CAR_{FC}$  in EP may reflect

the low OC content,  $< 4\%$  OC for most lakes in that region (Dong et al. 2012). Indeed, the relationship between  $MAR_{FC}$  and  $CAR_{FC}$  indicates that  $MAR_{FC}$  only explained about  $\sim 43\%$  of the  $CAR_{FC}$  change in EP (Fig. 5b), suggesting that the OC content may play a role in affecting OC burial rate. Several eutrophic lakes in EP exhibited low post-1950  $CAR_{FC}$  ( $< 30 \text{ g C m}^{-2} \text{ yr}^{-1}$ ), much lower than that of their counterparts elsewhere (Anderson et al. 2014). Although the primary production was typically high ( $> 100 \text{ g C m}^{-2} \text{ yr}^{-1}$ ), the low burial efficiency resulted in low OC accumulation for some lakes in this region (Dong et al. 2012). Lakes in EP are mostly large and shallow (Supporting Information Table S1), favorable for the aerobic decomposition of OM both suspended in the water and stored in the lake sediments (Wetzel 2001; Sobek et al. 2009). In addition, the high water temperature and nutrient condition of the EP lakes was suggested to be another possible factor that accelerates sediment OC mineralization (Dong et al. 2012). Indeed,

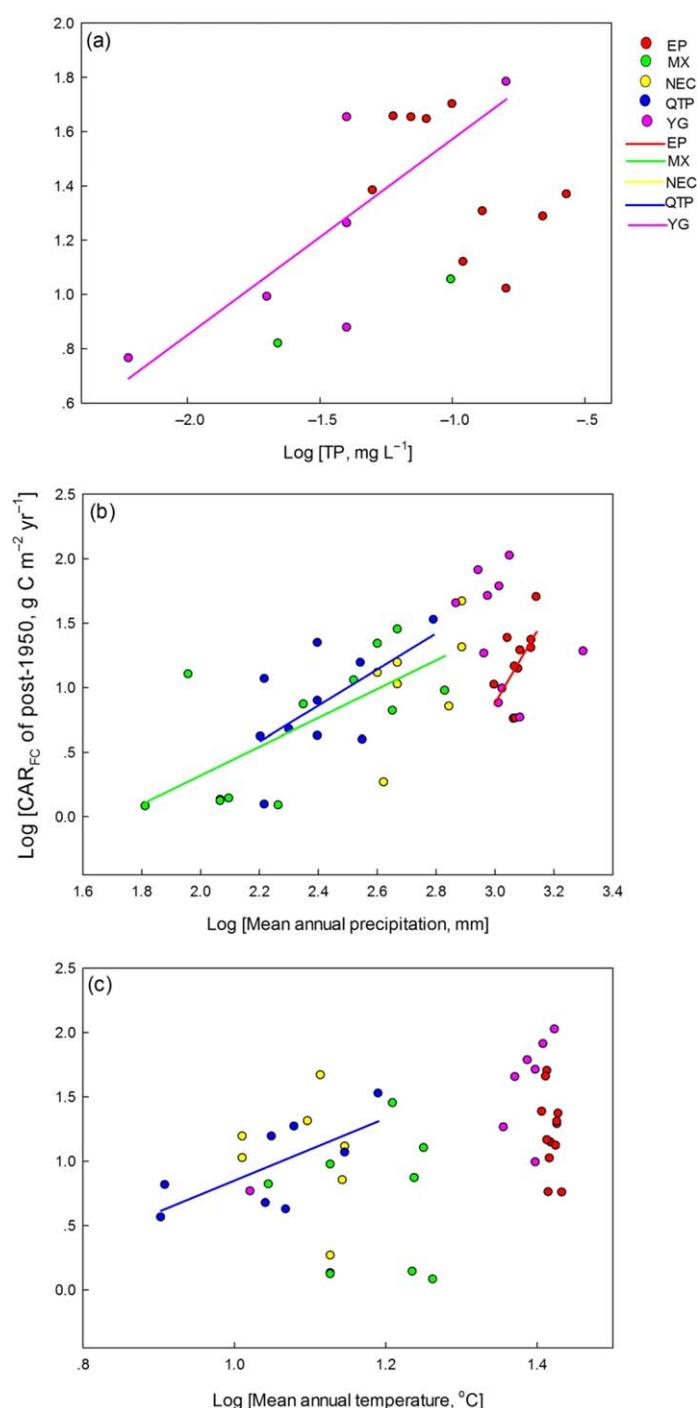


**Fig. 3.** (a) Mean change in OC burial rates during the three time periods for lakes in different regions. Error bars represent the 95% confidence interval around the mean and those that do not overlap zero represent a significant positive increase (*t*-test,  $p < 0.05$ ). (b) OC burial rate vs. time, binned by 20-yr intervals, from 1850 AD to 2000 AD. A LOWESS smoother (red line) was fitted to the median of all bins to show the general trend in the OC burial rate over time. [Color figure can be viewed at [wileyonlinelibrary.com](http://wileyonlinelibrary.com)]

post-1950  $CAR_{FC}$  was found to be negatively related to the regional mean annual air temperature (Table 2). Furthermore, although lakes in EP have intensive human activities in their catchments (Wang and Dou 1998), the impact of anthropogenic disturbance may be limited due to their large size.

#### Comparison with OC burial rate between lakes in other regions

The mean post-1950 lake  $CAR_{FC}$  in regions undergoing intensive anthropogenic disturbance in China was  $10.5 \pm 4.8$  g C m<sup>-2</sup> yr<sup>-1</sup> in EP, lower than those observed in



**Fig. 4.** The relationship between post-1950 OC burial rate and TP concentration (a), annual average precipitation (b), and annual average temperature (c) for lakes in each region of China. All data were log-transformed. [Color figure can be viewed at [wileyonlinelibrary.com](http://wileyonlinelibrary.com)]

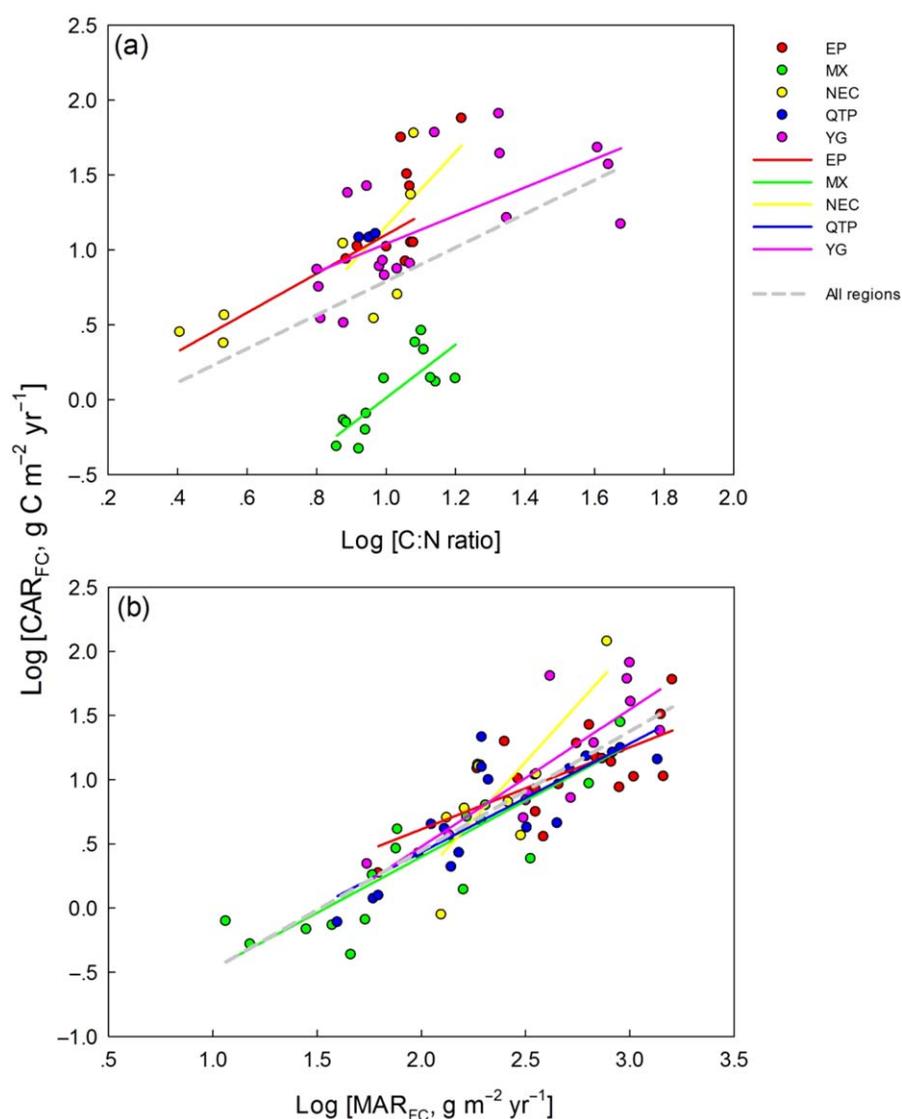
Europe ( $\sim 60 \text{ g C m}^{-2} \text{ yr}^{-1}$ ) (Anderson et al. 2014), the Conterminous United States ( $46 \text{ g C m}^{-2} \text{ yr}^{-1}$ ) (Clow et al. 2015) and agriculturally impacted lakes in central-southern Minnesota ( $39 \text{ g C m}^{-2} \text{ yr}^{-1}$ ) (Anderson et al. 2013). The lower

contemporary lake OC burial rate in EP may be due to their larger size (lakes mostly  $> 100 \text{ km}^2$ ; Supporting Information Table S1) when compared to those in other regions (generally  $< 5 \text{ km}^2$ ), which may have lessened the impact of anthropogenic disturbance on the OC burial rate. The contemporary OC burial rate for severely agriculturally impacted lakes in the U.S. was  $88 \text{ g C m}^{-2} \text{ yr}^{-1}$  (Heathcote and Downing 2012), higher than their counterparts in China and elsewhere, possibly resulting from a focusing correction not being applied. When comparing among lakes across regions, the effect of sediment focusing which leads to overestimates of the OC burial rate (Engstrom and Rose 2013; Anderson et al. 2014) must be considered. The highest lake  $\text{CAR}_{\text{FC}}$  ( $\sim 120 \text{ g C m}^{-2} \text{ yr}^{-1}$ ) observed in China was comparable to those rates observed in eutrophic lakes in Europe ( $\sim 200 \text{ g C m}^{-2} \text{ yr}^{-1}$ ) (Anderson et al. 2014) and North America ( $127\text{--}144 \text{ g C m}^{-2} \text{ yr}^{-1}$ ) (Heathcote and Downing 2012; Anderson et al. 2013). The mean  $\text{CAR}_{\text{FC}}$  in QTP where lakes are largely unaffected by anthropogenic activity ( $8.1 \pm 1.7 \text{ g C m}^{-2} \text{ yr}^{-1}$ ) was comparable to that observed in relatively undisturbed northern forest lakes ( $15 \pm 9.4 \text{ g C m}^{-2} \text{ yr}^{-1}$ ) (Heathcote et al. 2015).

#### Controls over contemporary OC burial in lakes of China

Lake OC burial rates are a function of the rate of primary production, the influx of terrestrial OM as well as post-deposition mineralization (Anderson et al. 2013, 2014; Wang et al. 2015). A warming climate may result in enhanced aquatic primary productivity (Zhang et al. 2012), because higher water temperature can increase microbial activity and internal nutrient cycling. A stronger effect of climate warming may be via its disruption of catchment hydrological budgets and hence delivery of both terrestrial carbon (as both dissolved and particulate OC) and nutrients (Schindler et al. 1996, 1997). Experimental studies suggest a negative effect of temperature on the OC burial rate in experiment treatments (Gudasz et al. 2010), but at the regional scale temperature exerts a weak but positive impact on lake OC burial (Heathcote et al. 2015; Wang et al. 2015). We found that the annual mean air temperature was significantly related to lake  $\text{CAR}_{\text{FC}}$  in QTP, MX, and NEC (Fig. 4c), suggesting that air temperature may influence aquatic primary production in the three northern/high altitude and colder regions but not in the southern warmer regions.

Precipitation has been shown to positively affect the OC burial rate by changing the terrestrial OM input and associated nutrient loading (Wang et al. 2015). In this study, precipitation was shown to exert a clear influence on  $\text{CAR}_{\text{FC}}$  for lakes across China, as well as regionally, with the exception of YG (Fig. 4b). However, lakes with high precipitation are also affected by intensive human activities in much of China, so the real effect of precipitation on lake  $\text{CAR}_{\text{FC}}$  can only be identified by excluding the influences of anthropogenic activity. In QTP, lakes have relatively limited human



**Fig. 5.** The relationship between (a) the post-1950  $\text{CAR}_{\text{FC}}$  and  $\text{MAR}_{\text{FC}}$ ; (b) the correlation between C : N ratio and  $\text{CAR}_{\text{FC}}$  of lakes in each region of China. All data were log-transformed. [Color figure can be viewed at [wileyonlinelibrary.com](http://wileyonlinelibrary.com)]

disturbance, making it possible to study the “real” effect of precipitation on lake  $\text{CAR}_{\text{FC}}$ . The strong relationship between lake  $\text{CAR}_{\text{FC}}$  and precipitation in QTP suggests that precipitation may be an important regulator of lake  $\text{CAR}_{\text{FC}}$  in undisturbed regions, in contrast to observations from lakes suffering intensive cultural disturbance (Anderson et al. 2013, 2014). This previous observation may be attributable to the different intensity of human disturbance among individual lakes, which can weaken or even outweigh the effect of climate on lake OC burial rates (Anderson et al. 2014). Indeed, an earlier study suggested that modern OC accumulation rate in Minnesotan lakes was primarily controlled by the intensification of agriculture in the catchment, not climate (Anderson et al. 2013).

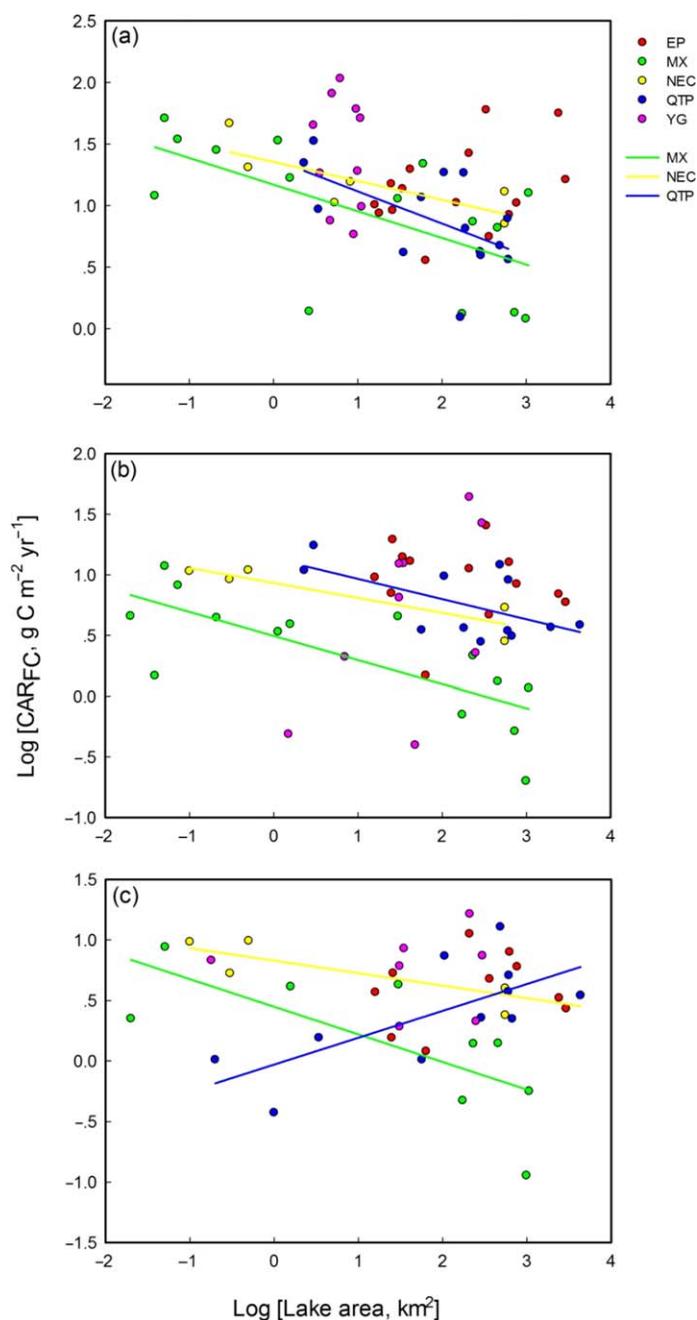
During the past century, many lakes in China have undergone nutrient enrichment by agricultural runoff, industrial, and domestic sewage disposal (Shang and Shang 2005). Nutrient enrichment can promote the in-lake primary productivity by promoting the development of nutrient-tolerant algae (e.g., increased diatom and cyanobacterial biomass) (Chen et al. 2011). About 50% of lakes in this study exhibited a rapid increase in  $\text{CAR}_{\text{FC}}$  after the 1970s or 1980s (Supporting Information Figs. S1–S3), partly attributable to the enhanced autogenic OM production due to the continuous increase of in-lake TP and the TN concentration (Chen et al. 2011; Gui et al. 2013). The contemporary lake  $\text{CAR}_{\text{FC}}$  was positively correlated with in-lake TP concentration in YG and the TN concentration in EP (Table 2). However, the

**Table 2.** Results of stepwise multivariate regression relating lake and catchment characteristics to the variability in  $CAR_{FC}$  for post-1950 for selected regions in China. Variables included in the model were lake surface area, in-lake TN, TP, mean annual air temperature ( $T$ ), mean annual precipitation ( $P$ ), pH, and water depth. All data were log-transformed and only significant ( $p < 0.05$ ) variables are shown. Regional abbreviations are as given in Table 1.

Region	Equation	$R^2$	$p$
EP	$Y = 47.8 + 0.87 \text{ TN} + 3.68 \text{ pH} + 6.78 P - 49.98 T$	0.76	0.01
MX	$Y = -7.72 + 1.24 P + 4.44 T$	0.39	0.02
QTP	$Y = -0.63 + 0.8 T + 0.27 P$	0.39	0.04
YG	$Y = 3.09 + 1.43 \text{ TP}$	0.82	0.03
NEC	$Y = -3.69 + 1.15 P + 1.35 T$	0.95	0.01

relationship between contemporary lake  $CAR_{FC}$  and TP was not significant in EP, probably due to the unreliable TP concentration data that were based on limited TP measurements from recent years and hence cannot be reliably compared with the decadal  $CAR_{FC}$  mean. Moreover, since lake  $CAR_{FC}$  represents not only the in-lake primary production, but also the processes of terrestrial OC input and OC degradation, the significant difference among individual lakes of these two processes could confound the effect of in-lake TP concentration on  $CAR_{FC}$ .

Since input of terrestrial OM can constitute an important component of the lake sediment OC content, it can influence the lake OC burial rate. Seston C : N ratio has been used as an indicator of historical changes in the source of lake sediment OM. Typically, algae have a C : N ratio between 4 and 10, whereas terrestrial OM has a C : N ratio greater than 20 (Meyers 1994). Therefore, increase in C : N ratio implies higher contribution of terrestrial OM to lake sediments. The regression analyses indicated that the sediment C : N ratio was positively linked to  $CAR_{FC}$  in all regions except for QTP ( $R^2 = 0.42\text{--}0.64$ ,  $p < 0.05$ ), suggesting that variation in terrestrial OM input was an important process explaining the spatial variability of lake OC burial in regions with strong catchment disturbance. During the past decades, many lakes have been affected by the extensive land reclamation for agriculture due to population growth (Chai et al. 2013; Gui et al. 2013). This anthropogenic activity destabilizes the topsoil, thereby aggravating soil erosion and allowing more terrestrial OM to enter the lakes. Indeed, for many lakes, trends in catchment land-use history were found to be similar to the temporal trend of  $CAR_{FC}$ . For example, the temporal trend in  $CAR_{FC}$  observed at many lakes since the 1950s can be attributed to the increasing extent of arable land in lake catchments (Dong et al. 2012; Gui et al. 2013). On the other hand, Gucheng Lake and Shijiu Lake, which are located in the same catchment, i.e., the



**Fig. 6.** The relationship between lake surface area and  $CAR_{FC}$  of lakes in each region of China for the period post-1950 (a), 1900–1950 (b), and 1850–1900 (c). All data were log-transformed. [Color figure can be viewed at [wileyonlinelibrary.com](http://wileyonlinelibrary.com)]

Qinyi river basin and Shiyang river basin (Xue et al. 2010; Xue and Yao 2011; Gui et al. 2013), have had a declining trend in  $CAR_{FC}$  since the early 1960s, probably due to the decrease in arable land cover (Gui et al. 2013). Over the last several decades, a growing number of reservoirs have been built for agricultural irrigation or flood control (Zhang et al. 2010), which significantly decreases the terrestrial OM inputs

**Table 3.** Estimation of total OC sequestration rate and OC storage by all lakes in different regions of China for the three time periods. Regional abbreviations are as given in Table 1.

Region	Total OC sequestration rate (Tg yr <sup>-1</sup> )			Total OC storage (Tg)		
	Post-1950	1900–1950	1850–1900	Post-1950	1900–1950	1850–1900
EP	0.38	0.25	0.11	19.21	12.44	5.33
YG	0.12	0.07	0.03	6.00	3.50	1.50
NEC	0.02	0.02	0.02	0.98	0.89	0.79
MX	0.03	0.02	0.02	1.44	1.03	0.80
QTP	0.33	0.24	0.17	16.74	11.80	8.38
Total	0.89	0.59	0.34	44.38	29.67	16.79

to downstream lakes. As a result of upstream reservoir construction, lakes, including Wushan (Zhang et al. 2010), Yue-liang (Chai et al. 2013), and Lianhuan (Sun et al. 2013), all exhibited declining CAR<sub>FC</sub> from the 1950s.

### Regional carbon budget implication

It is important to estimate the size and change of terrestrial OC stocks in order to develop strategies for mitigation of greenhouse gases. However, previous estimation of soil C dynamics ignored the lake C component in China (Xie et al. 2007; Piao et al. 2009). Xie et al. (2007) suggested a net soil C loss of 2.86 Pg, with the accumulation of 0.71 Pg in forest soils, but a loss of 3.55 Pg in grassland soils during the past 20 yr. The present study suggests that China's lakes buried ~ 0.05 Pg C since 1950 (Table 3), comparable to the C accumulation by forests but much lower than the soil C loss due to grassland conversion (Xie et al. 2007). Although the area of lakes is only 1/13 that of forests and 1/33 of grasslands in China, the contemporary lake OC burial rate was estimated as ~ 1 Tg yr<sup>-1</sup>, comparable to that of 4 Tg yr<sup>-1</sup> by forests and 6 Tg yr<sup>-1</sup> by grasslands (Piao et al. 2009), suggesting that lakes have considerable C sequestration capacity and cannot be ignored when studying the role of natural ecosystems in the regional C cycle.

This is the first estimation of focusing corrected OC accumulation dynamics of lakes in China at the national scale and helps our understanding of how contemporary OC burial rates vary with human activity across a range of climate zones (including monsoon-influenced regions) and provides an insight to the possible future fate of OC in lakes. However, our estimation can be biased due to the limited lake sediment <sup>210</sup>Pb data, and lack of bulk density data for several lake sediment cores. In addition, the site-specific atmospheric <sup>210</sup>Pb flux data were insufficient to generate accurate focusing-corrected CARs for all lakes. Moreover, the errors in the estimated mass and OC burial rates could also come from the uncertainties in the original source data themselves. Moreover, the estimation of limnic OC burial rates at regional and national scales can be biased by the limitations of up-scaling based on either a small dataset or variable regression relationships ( $R^2 = 0.35\text{--}0.82$ ) between

lake size and CAR<sub>FC</sub>. It is very difficult, however, to address all the possible sources of uncertainty and errors in the burial rate estimates presented in this study; given the range in rates observed among lakes within some regions, the real uncertainty may be underestimated. Therefore, addressing the knowledge and data gaps (i.e., increasing the number of lakes) is necessary to better understand of how climate and anthropogenic disturbance of biogeochemical cycles affects the dynamics of OC burial by lakes in China.

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#### Conflict of Interest

None declared.

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